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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of Takeshi Matsumoto et al.

Serial No.: 09/883,641

5 Filed: June 18, 2001

Title: EXHAUST GAS PURIFYING CATALYST AND METHOD FOR PURIFYING
EXHAUST GAS

Art Unit: 1754

Examiner: JONAS N. STRICKLAND

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COMMISSIONER FOR PATENTS

WASHINGTON, D. C. 20231

DECLARATION UNDER 37 C. F. R. 1.132

15 Sir:

I. I, Tatsuya YOSHIKAWA, of b-612, 1736-3, Imajuku, Himeji,
Hyogo 670-0052, Japan, hereby declare that I am one of the
inventors named in the above-mentioned application, that I
20 graduated from Kyoto University, Department of Engineering
and Petroleum Chemistry in March 1988 with a B.S. degree.
I received my master's degree, molecular engineering, from
the same University in March 1990, that, since April 1990,
I have been employed by Nippon Shokubai Co., Ltd., and engaged
25 in exhaust gas purifying catalyst research in the company.
In the period, I have been to Reading University, UK, for
2 years as guest researcher to study surface science from
1994 to 1996. Additionally, in accordance with our section
consolidation to ICT Co., Ltd, I have been in temporal transfer
30 to ICT Co., Since 1998.

II. I understand that claims 2, 18 and 19 are rejected under

35 U.S.C. 103(a) as being unpatentable over Kurokawa et al. (US Patent 5,958,826) in view of Kunio et al. (JP 07-047227), that claims 10 - 12, 14, and 15 are rejected under 35 U.S.C. 103(a) as being unpatentable over Kurokawa et al. in view of Horiuchi et al. (US Patent 5,911,961) and Farrauto et al. (US Patent 5,804,155), that claim 13 is rejected under 35 U.S.C. 103(a) as being unpatentable over Kurokawa et al. in view of Farrauto et al., and that claims 16 and 17 are rejected under 35 U.S.C. 103(a) as being unpatentable over Kurokawa et al. and Hidetoshi (JP 08-281111), and I have reviewed these references. It is of my opinion that the claimed catalysts in the present application are nonobvious over the cited references because the claimed catalyst achieved superior results as shown by the following experimental results.

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Experiment 1 (corresponding to Example 1 of the present specification)

A titania powder (available from Ishihara Sangyo Kaisha, Ltd. in Japan) was immersed with an aqueous dinitrodiamineplatinum solution, dried at 120°C, and then calcined at 500°C for one hour to obtain a Pt-deposited titania powder (Powder 1). The Pt concentration of this powder was 10 wt. % based on the weight of titania.

A quantity 0.22 g of the resultant Powder 1, 1000 g of a nickel oxide powder (reagent grade, available from Wako Pure Chemical Industries, Ltd. in Japan), 1080 g of a ZSM-5 type zeolite (available from Zeolyst International), and 2000 g of water were ball milled. In the produced slurry, an open flow type honeycomb carrier formed of cordierite (1.0 liter, 400 cells, available from NGK Insulators Ltd. in Japan) as a refractory carrier was immersed. Subsequently, the honeycomb carrier wet with the slurry was drained to expel

excess slurry and then dried at 150°C for two hours. The resulting catalyst element (Catalyst Element 1) contained 0.01 g of Pt, 0.1 g of TiO₂, 50 g of nickel oxide, and 50 g of ZSM-5, per liter of carrier.

- 5 Separately, 1 kg of an aqueous platinum ammine hydroxide solution (Pt: 1.2 g/liter) was mixed with 1200 g of a zeolite powder, stirred, then filtered and the resultant dried at 80°C for 12 hours, and subsequently calcined at 500°C for one hour to afford a Pt ion-exchange zeolite powder (Powder 2).
10 The Pt concentration of this powder was 0.1%.

- A slurry was formed by wet pulverizing 1092 g of Powder 2, 544 g of silica sol (solid content 20%) (available from Nissan Chemicals Industries Ltd. in Japan), and 1032 g of water. A honeycomb carrier (supra) was immersed in the
15 produced slurry. The honeycomb carrier wet with the slurry was drained to expel excess slurry and then dried at 150°C for two hours. The resulting catalyst element (Catalyst Element 2) contained 100.1 g of Pt-ZSM-5 and 10 g of SiO₂, per liter of carrier.

- 20 Disposing Catalyst Element 1 on the former stage (upstream side) and Catalyst Element 2 on the latter stage (downstream side) formed a catalyst.

Comparative Experiment 1

- 25 The procedure of Experiment 1 was repeated, except that the disposing order of the Catalyst Elements 1 and 2 were reversed. Thus, disposing Catalyst Element 1 on the latter stage and Catalyst Element 2 on the former state formed a catalyst.

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Experiment 2 (corresponding to Example 10 of the present specification)

The catalysts obtained in Experiment 1 and Comparative experiment 1 were tested for the ability to purify the exhaust gas by the following method.

5 In the test, a supercharging antechamber combustion type diesel engine (4 cylinders, 2.8 L) was used as the internal combustion engine, and light oil having a sulfur content of 0.05 wt. % used as the fuel for the internal combustion engine.

10 First, a given exhaust gas purifying catalyst was set at place in an exhaust gas tube connected to the diesel engine mentioned above and a temperature elevation test from 150°C to 350°C (20°C/min) was carried out by a programmed operation. The programmed operation was performed by the use of an automatic engine-operating device (available from Horiba Seisakusho Ltd. in Japan and sold under the product code of
15 "HERT-381" under the condition of 1500 rpm), with the magnitude of automatic torque varied.

Then, the programmed operation was performed in the absence of the catalyst from the exhaust gas tube. The gas then exhausted from the engine was continuously sampled and
20 analyzed with a continuous gas analyzer to determine NO_x and hydrocarbon contents in the exhaust gas. NO_x was analyzed with a chemical emission spectral analyzer (CLD) and the hydrocarbon with a hydrogen flame ionizing analyzer (NDIR) respectively. The analysis results at various temperatures
25 are shown in Table 1.

TABLE 1

Gas temperature	150° C	175°C	200°C	250°C	300°C	350°C
Rotational frequency (rpm)	1500	←	←	←	←	←
THC (ppm)	68	60	57	55	35	32
NOx (ppm)	70	72	82	83	85	95
THC/NOx	0.97	0.83	0.70	0.66	0.41	0.34

Then, the exhaust gas that had contact with the catalyst was analyzed to determine NO_x and hydrocarbon contents. The purification ratios (the degrees of conversion) of NO_x and hydrocarbon were calculated based on the contents of NO_x and hydrocarbon in the exhaust gas determined in the absence of the catalyst from the exhaust gas tube and the contents of NO_x and hydrocarbon determined in the presence of the catalyst in the exhaust gas tube. Let XO (mol) stand for the content in the exhaust gas in the absence of the catalyst and X1 for the content in the presence of the catalyst, the calculation will be attained by the following formula:

$$\text{Purification ratio (degree of conversion) (\%)} = \frac{(XO - X1)}{XO} \times 100 .$$

The capacity for purifying NO_x (degree of purification) at various temperatures found by the calculation is shown in Table A, and Fig. A attached.

TABLE A

	150°C	175°C	200°C	250°C	300°C	350°C
Experiment 1	38	30	35	29	12	10
Comparative Experiment 1	35	28	25	-25	-3	7

CONCLUSION

It is clear from the above Table A that the catalyst of the present invention provided superior NOx conversion to the catalyst of the Comparative Experiment 1.

III. The undersigned declares that all statements made herein are true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fines or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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2003/10/22

Date

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